Health Physics Vol. 41 (July), pp. 59-67, 1981
Printed in the U.S.A.

0017-9078/81/070059-09$02.00/0
Pergamon Press Ltd.

DECREASE OF RADIONUCLIDES IN COLUMBIA RIVER BIOTA FOLLOWING CLOSURE OF HANFORD REACTORS

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(*Received* 24 *March* 1980; *accepted* 21 *October* 1980)

Abstract—In January 1971, the last of nine plutonium production reactors using direct discharge of once-through cooling waters into the Columbia River was closed. Sampling was conducted at three stations on the Columbia River to document the decline of radionuclides in the biota of the Columbia River ecosystem.

Concentrations of ^{60}Co in seston, periphyton and invertebrates did not decrease to the degree that the other radionuclides did; this is partially related to the seepage of ^{60}Co into the river from a disposal trench near the operating *N* Reactor. Levels of ^{60}Co in fish showed some decreases, but obvious trends were not present. Zinc-65 was present in the biota in highest concentrations. The amounts in seston and periphyton decreased rapidly and were measurable only until the spring of 1973. Zinc-65 in caddisfly larvae was not measurable by February 1973, but concentrations in McNary chironomids fluctuated between unmeasurable levels to 24 pCi/g dry weight (DW) and this was related to ingestion of contaminated sediments rather than larval concentrations. In suckers and squawfish, ^{65}Zn decreased to fairly low, constant levels of 1 and 3 pCi/g DW, respectively.

The data show that in a river-reservoir complex, the measurable concentrations of fission-produced radionuclides decreased to extremely low or unmeasurable levels within 18–24 months after cessation of discharge of reactor once-through cooling water into the river. On the basis of data from the free-flowing sampling station, we believe that the decrease would be even more rapid in an unimpounded river.

INTRODUCTION

In 1944, the first plutonium production reactor on the Atomic Energy Commission's Hanford Reservation (now the Dept. of Energy's Hanford Site) began discharging radionuclides into the Columbia River via the cooling water effluents. In succeeding years, as many as nine reactors released radionuclides into the Columbia at one time. With the closure of the K East Reactor in January 1971, direct discharge of once-through cooling water into the Columbia ended. The only radioactivity presently reaching the river is from N Reactor coolant water which is periodically discharged into a seepage trench. From this trench, small amounts of the coolant water seep through the ground to the river (Ro73).

A large body of literature, abstracted by Becker (Be73), has reported the results of radioecological studies of the Columbia River, the most thorough surveys being those of Robeck *et al.* (Ro54), Davis *et al.* (Da56) and Watson *et al.* (Wa70). Watson *et al.* (Wa69) also studied the changing concentrations of radionuclides in Columbia River biota during a 40-day closure of all reactors in 1966. Numerous studies reported the accumulation and loss of radionuclides by biota, but the closure of the Hanford reactors provided a unique opportunity to study the pattern of decreasing radionuclide concentrations in the components of the natural river ecosystem over time.

Knowledge of the loss of radionuclides from biota is particularly important today, with the number of nuclear power generating plants increasing. Although reactors currently being built do not discharge the same suite of radionuclides that the Pu production reactors did, the data reported here may ensure a more complete evaluation of the environmental impact of siting nuclear facilities. The data are also applicable to the release to flowing waters of radionuclides from other sources, such as fuel-reprocessing plants and rad-waste disposal sites, and from the decommissioning of facilities.

The purpose of this investigation was to determine the decline in concentration of radionuclides in the biota of the Columbia River ecosystem after shutdown of the Hanford reactors with once-through cooling systems. Cobalt-60 and ^{65}Zn are emphasized because these radionuclides were present in sufficient amounts to provide a meaningful discussion. Scandium-46, ^{54}Mn and ^{137}Cs were also measured, but values were erratic; a brief discussion of their concentrations is given.

SAMPLING LOCATIONS

Three sites were sampled from July 1971 through June 1972 (Fig. 1), and one site, McNary Reservoir, was sampled until June 1973. Physical descriptions of the stations can be found in Cushing *et al.* (Cu80).

METHODS AND MATERIALS

Biota sampled

Initially, we sampled seston, periphyton, a dominant (by numbers and biomass) invertebrate [caddisfly larvae (*Hydropsychidae*) at White Bluffs, chironomids (*Chironomidae*) at McNary], an herbivorous fish species (sucker, *Catostomus macrocheilus*), and a carnivorous fish species (squawfish, *Ptychocheilus oregonensis*) at each site. These organisms are representative of the several trophic levels and nutrient pathways.

Field collection

Seston was collected by towing a plankton net (Nitex®, 80 μm mesh). Periphyton was collected mainly from artificial substrates because power production by the dams resulted in water level fluctuations such that accessible natural substrates were not always submerged.

Caddisfly larvae at White Bluffs were collected by hand-picking from rocks in riffle areas. Chironomids in McNary Reservoir

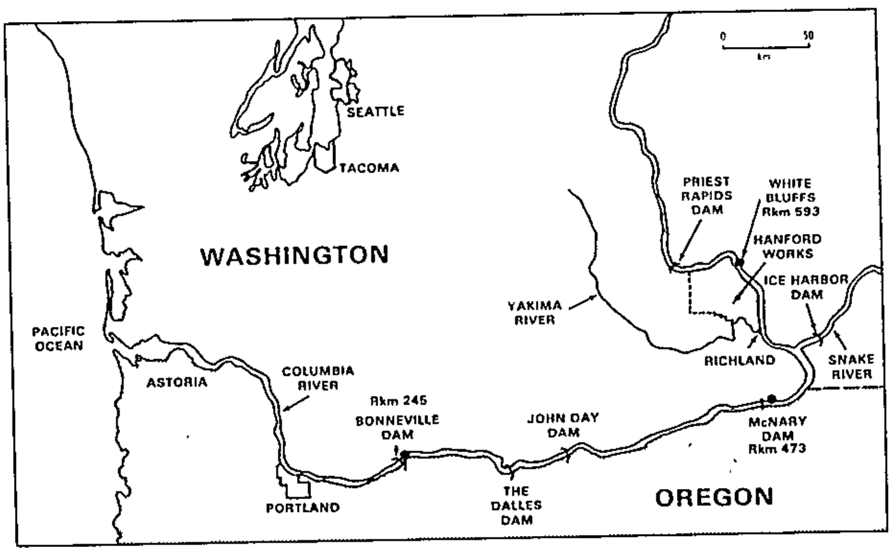

FIG. 1. Location (dots) of sampling stations.

were collected by washing sediments through screens.

Suckers and squawfish were collected with 46-m (125 ft) long experimental gill nets [five mesh sizes from 2.54 cm (1 in.) to 7.62 cm (3 in.)].

Laboratory processing

Samples were prepared for γ-ray spectrometry by filling a counting container (10 cm in diameter by 2 cm thick) with an homogenized, ashed and weighed aliquot. If sufficient tissue was not present to fill the container, the entire sampled was uniformly mixed with agar–agar to make up the difference. The gut contents of suckers and squawfish were removed before processing. Prior to ashing, samples were oven-dried at 60°C and weighed. The number of samples of each type counted varied from 1 to 4, and in some cases for the larger fish, the single analysis was from a composite sample of up to 10 individual fish. All $\pm$ values refer to 1 S.D. of the mean and refer to biological, not counting, variability. The samples were counted on an anticoincidence shielded, multidimensional γ-ray spectrometer (Wo67). Minimum detection limits (MDL) for ^{46}Sc and ^{60}Co in seston, periphyton, suckers and squawfish were 0.001 pCi/g DW. MDL for ^{46}Sc in caddisfly and chironomid larvae, and ^{54}Mn, ^{65}Zn and ^{137}Cs in seston, periphyton, suckers and squawfish was 0.01 pCi/g DW. MDL for ^{54}Mn, ^{60}Co, ^{65}Zn, and ^{137}Cs in caddisfly and chironomid larvae was 1.0 pCi/g DW.

Sampling for this study did not begin until June 1971, about 5 months after closure of the last once-through cooling reactor. Thus, some unknown decrease in radionuclide concentration in the biota occurred before our sampling began. This prevents the calculation of accurate rates of decline or effective half-lives from the time all reactors were operating. However, to provide an indication of the concentrations of these radionuclides in the biota while five reactors were in operation, the data in Table 1 (Wa70) are presented. These data were obtained from organisms collected just downstream from the White Bluffs area and can be compared with values presented in the text and figures. An additional factor to be considered is that between 1967, when the last sampling was done in the studies by Watson *et al.* (Wa70), and January 1971, when the last reactor was closed, the four other reactors were shut down sequentially, so that the unknown loss of radioactivity in the organisms in the interim was also probably of a "stair-step" nature.

RESULTS AND DISCUSSION

It should be kept in mind that the decreasing concentrations of the radionuclides in the biota described below are a result of three coincident processes: (1) physical decay of the radionuclide, (2) biological turnover of

Table 1. Radioactivity in selected Columbia River biota, 1966 and 1967 (modified from Wa70)

Organism	Number Sampled	pCi/g DW ^{60}Co	pCi/g DW ^{65}Zn
Seston	45	52 $\pm$ 201[a]	10720 $\pm$ 8290
Periphyton	35	240 $\pm$ 480[a]	8190 $\pm$ 5940
Caddisfly larvae	39	66 $\pm$ 104[a]	3686 $\pm$ 2186
Suckers	5	1.4 $\pm$ 0.2	249 $\pm$ 62
Squawfish	2	0.5 $\pm$ 0.6	141 $\pm$ 77

[a] 50% or more of values below detection limits, but included in calculation of means as a zero value.

the element by the organisms, and (3) decreasing radionuclide availability in the water and food supply (in most cases). In this study, the third item is a function of the first and second. Since the study period included 3.0 half-lives for ^{65}Zn, the decline due to physical decay for this isotope is significant; that for ^{60}Co is negligible because of its long half-life.

Cobalt-60 (*half-life*, 5.24 yr)

Concentrations of ^{60}Co in seston at White Bluffs fluctuated during the first year with no appreciable overall decline. This was probably related to the proximity of the N Reactor (15 km upstream), from which ^{60}Co leaches into the Columbia River through springs draining a disposal trench (D. E. Robertson, personal communication; Cushing and Watson, unpublished data). Mean concentration at White Bluffs was 6.0 ± 3.3 pCi/g DW ($n = 12$). An overall decrease from about 20 pCi/g DW to values near unity was measured over the 2 yr in seston at McNary (Fig. 2a). Concentrations of ^{60}Co in seston at Bonneville also decreased during the first year from about 5 to 1 pCi/g DW (Fig. 2a).

Cobalt-60 concentrations in periphyton at White Bluffs decreased from 22 to 2 pCi/g DW during the first year of the study (Fig. 2b). In McNary, concentrations decreased from 34 to about 3 pCi/g DW during the first year and fluctuated between about 1 and 7 pCi/g DW during the second year. At Bonneville, concentrations declined from about 5 to 1 pCi/g DW (Fig. 2b).

Caddisfly larvae at White Bluffs showed no appreciable decline of ^{60}Co during the 6 months they were available. Mean concentration was 12.0 ± 2.5 pCi/g DW ($n = 6$). The pattern of ^{60}Co concentrtion in chironomids in McNary was unusual (Fig. 2c); concentrations increased from 5.4 to 28.2 pCi/g DW from June to October 1972 and then decreased and remained essentially constant at about 5 pCi/g DW for the duration of the study.

There is some evidence of a decrease in ^{60}Co concentration in suckers from White Bluffs (Fig. 2d), although the fluctuations at the beginning and end of the first year overlap. Mean concentration was 0.68 ± 0.5 pCi/g DW ($n = 13$). Cobalt-60 concentrations in suckers in McNary decreased from about 0.39 to 0.06 pCi/g DW during the 2 yr of study (Fig. 2d). No obvious trend in ^{60}Co concentrations in suckers from Bonneville was found (Fig. 2d). Cobalt-60 concentrations in squawfish at White Bluffs exhibited alternating increases and decreases (Fig. 2e). Cobalt-60 concentrations in squawfish from McNary showed no overall trend; mean concentration was 0.08 ± 0.1 pCi/g DW ($n = 27$) (Fig. 2e). At Bonneville, concentrations in squawfish decreased from 0.12 to 0.03 pCi/g DW in the first 3 months of study and remained essentially constant for the next 11 months (Fig. 2e).

Zinc-65 (*half-life*, 245 *days*)

In general, ^{65}Zn was the radionuclide in highest concentration in the biota. Concentrations of ^{65}Zn in seston at White Bluffs decreased quite rapidly, as would be expected of this transient community (Fig. 3a). After the initial decrease from about 38 to 3 pCi/g DW, ^{65}Zn levels fluctuated during the following year. The increase of ^{65}Zn in April 1972 in this and other organisms coincided with the spring runoff, a period in which ^{65}Zn sorbed to fine sediments is scoured and resuspended in the water column. This could make this radionuclide available for biological uptake. Zinc-65 in McNary seston did not decline as much, initially, as it did at White Bluffs, nor did it decrease as far until about March 1972 (Fig. 3a). At Bonneville, seston ^{65}Zn concentrations fluctuated similarly as at White Bluffs (Fig. 3a). At the end of the first year of study, ^{65}Zn concentrations were about 3 pCi/g DW at all sites. Zinc-65 concentrations in seston at McNary gradually decreased during the second year of study, with some fluctuations, to essentially unmeasurable levels (Fig. 3a). Decreases of ^{65}Zn in periphyton were similar to decreases in the seston (Fig. 3b). The April 1972 pulse was evident at White Bluffs, but it was less evident at McNary and absent at Bonneville. Cushing and Watson (Cu71) estimated an effective half-life of 15 days for ^{65}Zn in periphyton in laboratory streams.

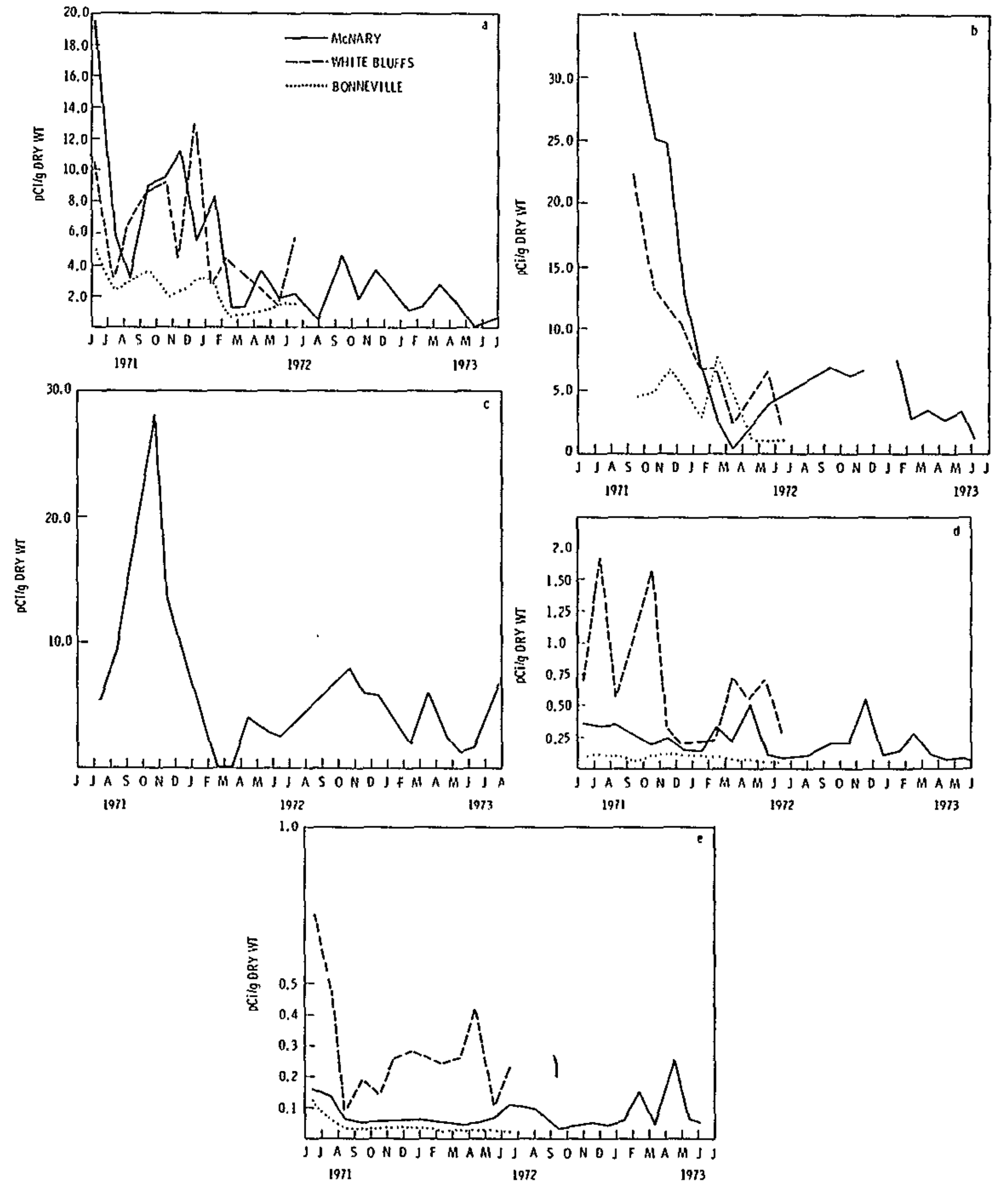

FIG. 2. Concentrations of ^{60}Co in selected Columbia River biota; (a) seston, (b) periphyton, (c) chironomids, (d) suckers, (e) squawfish.

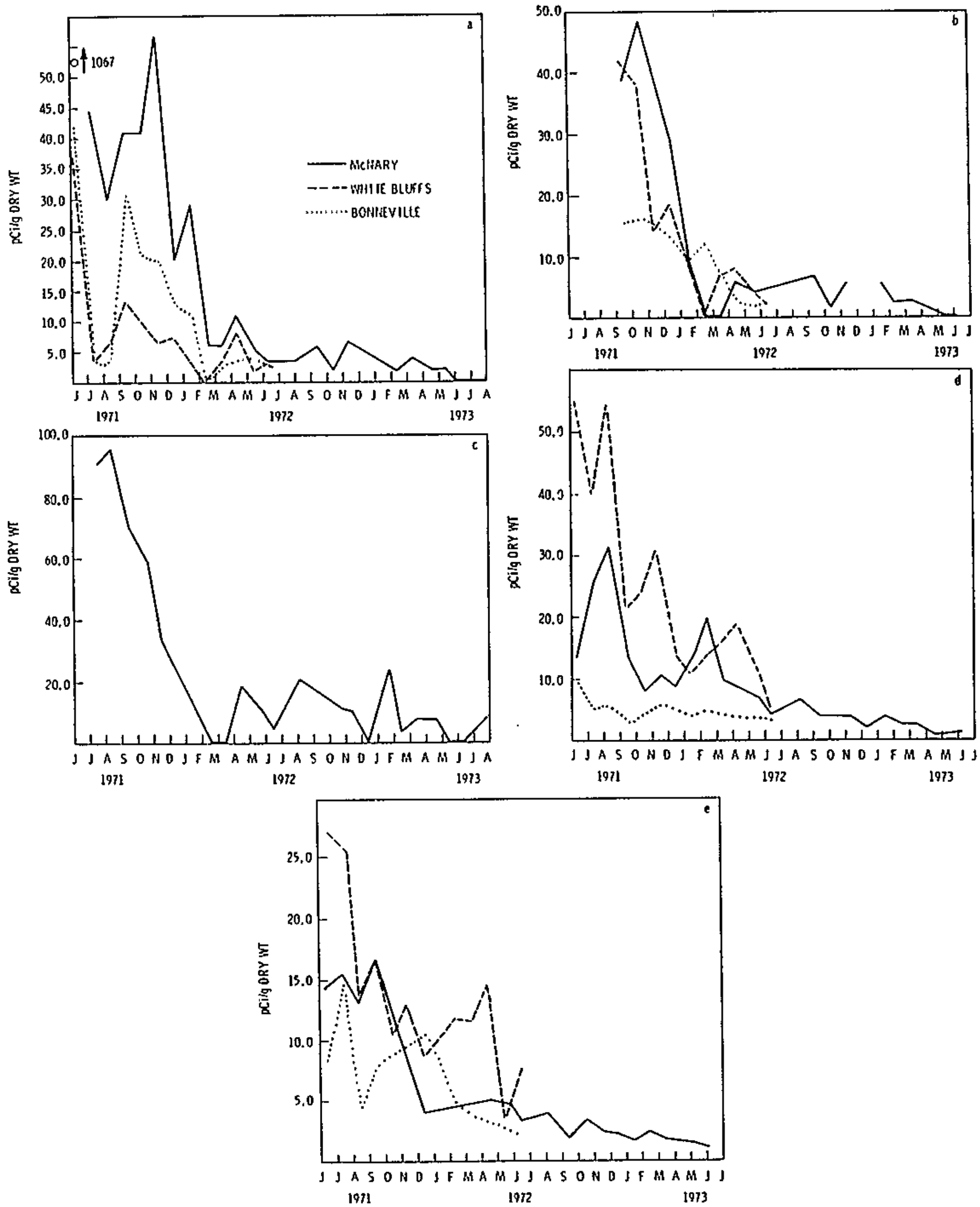

FIG. 3. Concentrations of ^{65}Zn in selected Columbia River biota; (a) seston, (b) periphyton, (c) chironomids, (d) suckers, (e) squawfish.

The concentration of ^{65}Zn in caddisfly larvae at White Bluffs decreased from about 11 pCi/g DW in September 1972 to unmeasurable levels by February 1973. Zinc-65 concentrations in McNary chironomids decreased rapidly from about 95 to less than 10 pCi/g DW in 6 months (Fig. 3c). Concentrations fluctuated between unmeasurable amounts to 24 pCi/g DW from February 1972 to the end of the study. Dean (De74) found that tubificid worms, which ingest sediments as do chironomids, accumulated ^{65}Zn from the water but not from the sediments. Thus, the decreasing concentrations of ^{65}Zn in chironomids may be partially related to decreased concentration in the water rather than the sediments. Renfro (Re73), on the other hand, found that marine polychaete worms accumulated ^{65}Zn from radioactively labelled sediments. In the same study, it was demonstrated that the burrowing activities of the polychaetes removed considerable ^{65}Zn from the sediments (Re73). Although decreasing sediment activity at McNary is related more to resuspension and transport and/or burying by new uncontaminated sediments, loss of ^{65}Zn due to agitation by chironomid larvae may be a contributing factor.

Zinc-65 concentrations in suckers at White Bluffs decreased from about 55 to 5 pCi/g DW during the first year of study (Fig. 3d). At McNary, concentrations fluctuated during the first year and then gradually decreased during the second year to values of about 1 pCi/g DW (Fig. 3d). Lower values were present at Bonneville; they decreased and remained fairly constant at about 3 pCi/g DW by the end of the first year (Fig. 3d). Concentrations of ^{65}Zn in the carnivorous squawfish at White Bluffs fluctuated but showed an overall decrease from about 27 to 5 pCi/g DW during the first year (Fig. 3e). At McNary, the rapid decrease during the first year slowed during the second; concentrations at the conclusion of the study were about 1.5 pCi/g DW (Fig. 3e). Concentrations also fluctuated at Bonneville, but revealed an overall decline during the first year (Fig. 3e). Jones (Jo75) calculated ecological half-lives (defined as the time the organism requires to lose 50% of the body burden by both decay and biological turnover = effective half-life) of ^{65}Zn in various tissues from carp in McNary at the same time our study was in progress. He found no statistical difference among the values for the various tissues and so derived a common loss rate with an ecological half-life of 177 days.

Analyses of water samples showed that about 20% of the ^{65}Zn was in ionic form when the reactors were operating (Pe66), but that less than 2% was ionic in 1971 and 1972 (Ro73), a tenfold decrease. With these data in mind, it is of interest to compare ratios of ^{65}Zn in various organisms and in their principal food during the above periods of time. Suckers graze on periphyton communities; when the reactors were operating, there was approx. 10 times as much ^{65}Zn in the periphyton as in the suckers (Wa70). In 1971 and 1972, there was only about 2 times as much ^{65}Zn in the periphyton. The same ratios prevailed for the squawfish, which prey on smaller forage fish. The change in ratio approximated that of the changes in ionic ^{65}Zn. The decrease of ^{65}Zn by 9–10 times in the chironomids closely approximates the tenfold decrease of ^{65}Zn in the sediments. Since chironomids ingest sediments, the decrease essentially indicates that radionuclide measurements of chironomids are reflective of the gut contents rather than the tissue.

Other radionuclides

Concentrations of ^{46}Sc (half-life, 84 days) decreased to unmeasurable levels in most biota by spring of 1972; large fish averaged about 0.03 pCi/g DW and also had unmeasurable levels after 1 yr. Concentrations of ^{54}Mn (half-life, 290 days) in the lower trophic levels were essentially unmeasurable after 1 yr. In suckers, concentrations declined to fairly constant levels of < 1 pCi/g DW at White Bluffs, a value similar to those found at all times at McNary Reservoir and Bonneville Reservoir. Values in squawfish became unmeasurable in the last year of the study. Levels of ^{137}Cs (half-life, 30 yr) were unmeasurable initially in periphyton, but measurable levels in the lower trophic levels were sporadic. Cesium-137 levels in suckers fluctuated markedly with no long term decrease. In squawfish, ^{137}Cs levels fluctuated at White Bluffs but were constant at McNary and Bonneville. See Cushing *et al.* (Cu80) for a fuller discussion of the variations in concentrations of ^{46}Sc, ^{54}Mn and ^{137}Cs during this study.

CONCLUSIONS

The biota in the Columbia River ecosystem below Hanford are being exposed to a changing, and much lower, ambient level of radionuclides in their environment. True effective half-lives of the radionuclides cannot be determined since radionuclides are still available from the sediments, from N Reactor seepage effluents, and from the residual radioactivity in the different organisms in the food web. The large sediment-bound radionuclide pool in McNary Reservoir will become less available to biota with time as it is covered with successive layers of sediments carrying only radionuclides related to the N Reactor, a much smaller fraction than originally found.

Nevertheless, these data show that in a river-reservoir complex, the measurable body burden of fission-produced radionuclides decreased to essentially unmeasurable levels within 18 to 24 months of cessation of input of once-through cooling water into the river. We further hypothesize, on the basis of the data from the White Bluffs station, that the decline would be more rapid, particularly in the lower trophic levels, in a free-flowing river. The current would transport the smaller constituents (e.g. seston, fine sediments) downstream, thus eliminating this pool containing the highest levels of radioactivity for recycling. Slack water allows the sediments and their associated radionuclide burden to settle and accumulate, thus providing a pool for subsequent resuspension and recycling of radionuclides within the food web.

Acknowledgements—We would like to thank R. L. Burdette and others for their capable assistance in the field and laboratory. The work was performed under Contract EY-76-C-06-1830 between the Dept. of Energy and Pacific Northwest Laboratory, operated by Battelle Memorial Institute. Funding for publication was provided by the Radioecology of Nuclear Fuel Cycles Project supported by the Office of Health and Environmental Research, Dept. of Energy.

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